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AN IMPROVED TV-SCANNING METHOD FOR RECORDING TIME-RESOLVED OPTICAL SPECTRA OF TRANSIENTS

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K. H. Schmidt, S. Gordon,
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ARGONNE NATIONAL LABORATORY, ARGONNE, ILLINOIS

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ARGONNE NATIONAL LABORATORY
9700 South Cass Avenue
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March 1976

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ABSTRACT

An improved method to record a time-resolved transient absorption or emission spectrum produced by a single pulse of radiation is described. A streak image of the spectrum is produced by an image-converter camera. The image is scanned by a TV camera, stored in a video-disk recorder, and transferred line by line to a computer. The computer can produce three-dimensional plots of absorbance or emission versus time and wavelength containing 100 x 100 data points.

I. INTRODUCTION

In a previous publication[1] we described a new method of recording the time dependence of a transient absorption or emission spectrum produced by a single pulse of radiation. The time-resolved spectrum is produced by an image-converter camera with streak capability. In the earlier publication we described the method in which the streak image was scanned by a TV camera, the output of which was transferred to a computer. In that method, 2000 data points were divided between wavelength and time points in different possible combinations.

In the improved version described in this report, we have increased the number of data points to 10,000 (100 wavelength and 100 time points). We have accomplished this by recording the TV output of the streak image onto a video disk recorder and transferring this image to the computer after digitizing. The data are processed by the computer and presented in a three-dimensional format on a Tektronix graphic terminal equipped with a hard-copy unit. Increased linearity is achieved by improving the electronic circuitry; increased sensitivity by use of a more sensitive Vidicon tube has also been achieved.

II. DESCRIPTION OF APPARATUS

The following description assumes familiarity with the apparatus described in Ref. 1. It includes only those features of the present system that have been added to or represent modifications of the previous version.

A. Optical Arrangement

The optical system has remained essentially the same as the one described in Ref. 1. Figure 1 shows our present arrangement. The following modifications may be noted.

1. The analyzing light makes only one pass (5 cm) through the absorption cell. Although this change reduces the absorption signal, the S/N ratio, especially in the far uv, is improved, because we can now make full use of the aperture of the spectrograph, and one mirror with its reflection losses has been eliminated.

2. A cylindrical lens has been inserted before the spectrograph to create a slight astigmatism, so that the "vertical" image of the arc appears on the vertical entrance slit of the spectrograph and the horizontal image on the horizontal slit, which is about 2 cm before the vertical slit. Due to the astigmatism inherent to the Czerny-Turner-Optics of the spectrograph, the images of both slits impinge on the photocathode of the spectrograph. This modification increases the light flux by a factor of 2-3.

3. A TV camera (Shibaden 16S) equipped with a Silicon Diode Array Vidicon (RCA 4532A) is now used. The new tube is about 40 times as sensitive as the standard Vidicon tube (RCA 773S).

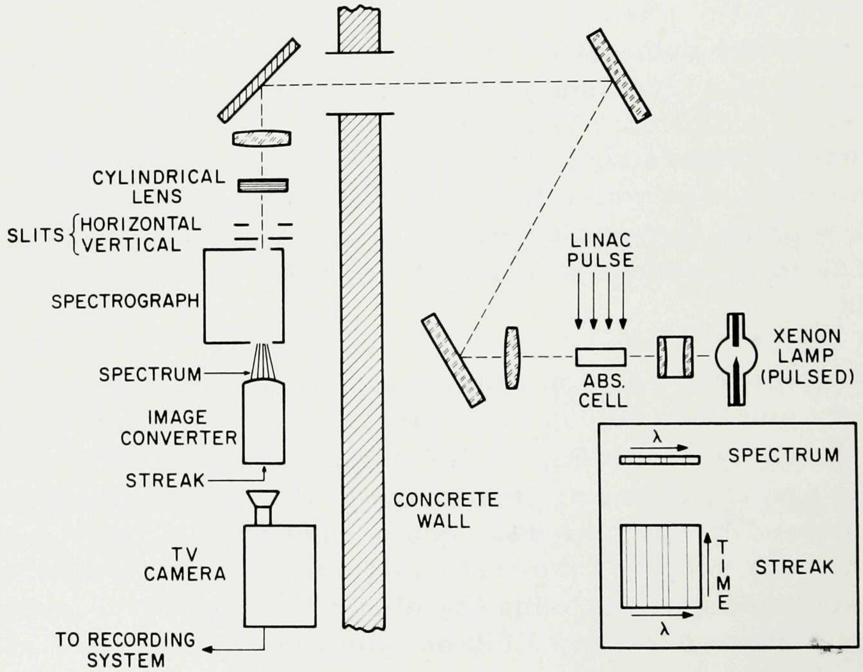


Fig. 1. Optical Arrangement of the Apparatus. ANL Neg. No. 122-1229.

B. Electronic Circuitry

The essential feature of the new streak apparatus is its capability to record and process the entire transient spectrum by storing the TV image of the streak on a TV disk recorder. We are using a relatively inexpensive Hitachi Model MV-1U ("Memory Vision") recorder, which stores a single TV field (263 lines) on a 10-cm magnetic disk.

Figure 2 shows the block diagram of the entire system. It is divided into two separate sections: The recording section stores a streak image on the video disk; the playback section transfers this data into the computer.

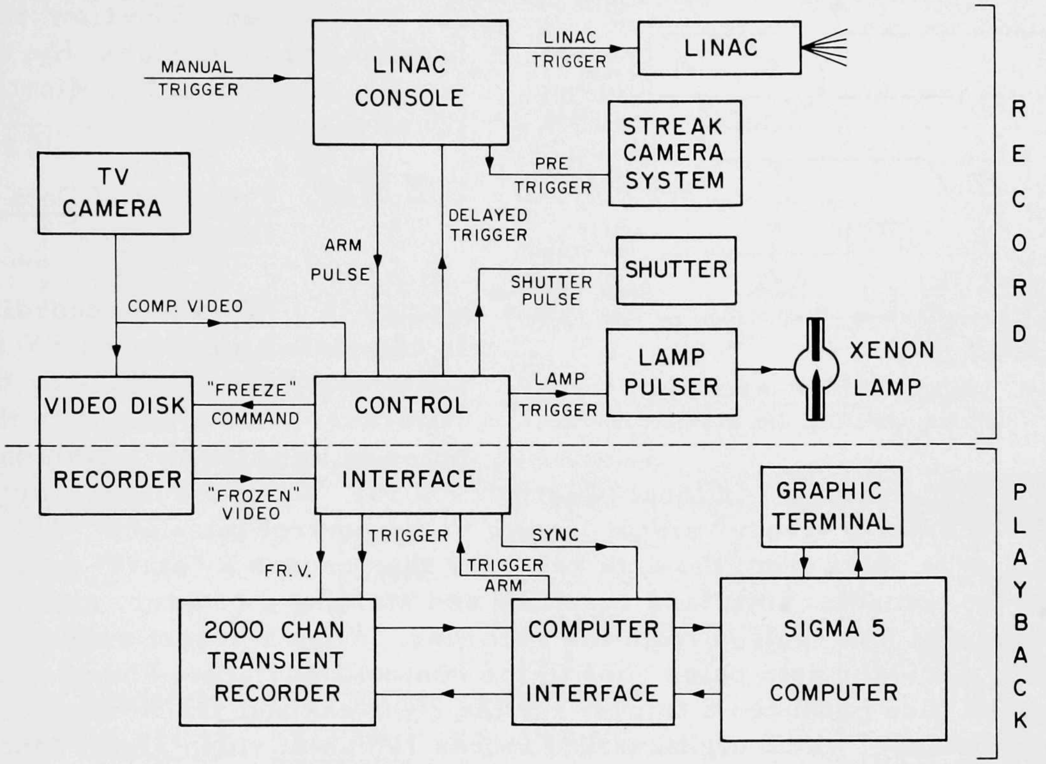


Fig. 2. Block Diagram of the Electronic Circuitry. ANL Neg. No. 122-1228.

1. Recording Sequence

The sequence of events of the recording process, which is schematically indicated in Fig. 3, is as follows: A manually initiated trigger signal is sent to the linac console and arms the linac. After a variable time, the console returns a pulse to the control interface. This pulse arms a circuit which releases a trigger pulse when the next vertical sync pulse from the TV camera preceding an even-numbered line scan arrives. In Fig. 3 this is the first sync pulse labeled 2.

All further events are in a fixed time relation to this trigger: A "freeze" command is sent to the video recorder, about halfway between two vertical sync pulses. The following vertical sync pulse then starts an "erase" period of 1/60 s, and 1/30 s later ("time zero"), a recording period (1/60 s). An electronically controlled shutter opens 5 ms before time zero and closes 5 ms after time zero. Also at time zero, the xenon lamp pulser is triggered, and about 0.2 ms later, a trigger is sent to the linac console. After transmitting a pretrigger to the streak camera system, the console fires the linac.

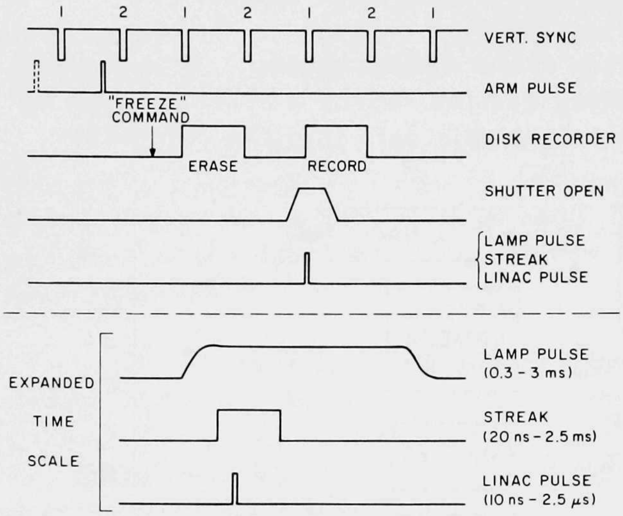


Fig. 3. Sequence of Events in Recording a Streak Image. ANL Neg. No. 122-1230 Rev. 2.

By means of the delay generator included in the streak camera system (not shown in the figure) the position of the linac pulse relative to the beginning of the streak can be adjusted. The time relation between the lamp pulse, the streak, and the linac pulse is indicated in the lower part of Fig. 3.

2. Transfer of Data to Computer

After the recording phase is completed, the stored TV field is continuously played back by the video recorder. Data transfer to the computer is initiated by a command typed on the graphic display terminal (Tektronix 4010). When the computer is ready to receive data, a "ready" signal is sent to the control interface. The next vertical sync pulse from the disk recorder then causes a "start" pulse to be sent to the computer interface resetting and starting a counter, which counts the horizontal sync pulses from the recorder. When a preset number is reached, a trigger arm pulse goes to the control interface. The next horizontal sync pulse then produces a trigger for the 2000-channel transient recorder (Biomation 8100) which digitizes and stores 100 μs of video signal (one TV line plus parts of the two adjacent lines). The digitized data are automatically transferred to the computer disk.

The subsequent "ready" signal increments the preset value for the counter by one, and the same sequence is repeated, thus transferring the next TV line. This process continues until the desired number of lines are stored on the computer disk. The numbers of the first and last TV lines to be transmitted can be entered initially through the graphic terminal. Usually 100 lines (about the height of the streak image) are used.

3. Recording and Playback Logic

Figure 4 shows the block diagram of the control interface. Detailed circuit diagrams for the "Record" section are given in Figs. 5 and 6, diagrams for the "Playback" section in Figs. 7 and 8. Not included in these figures are the blocks labeled "Modulator" and "Demodulator," which will be described separately.

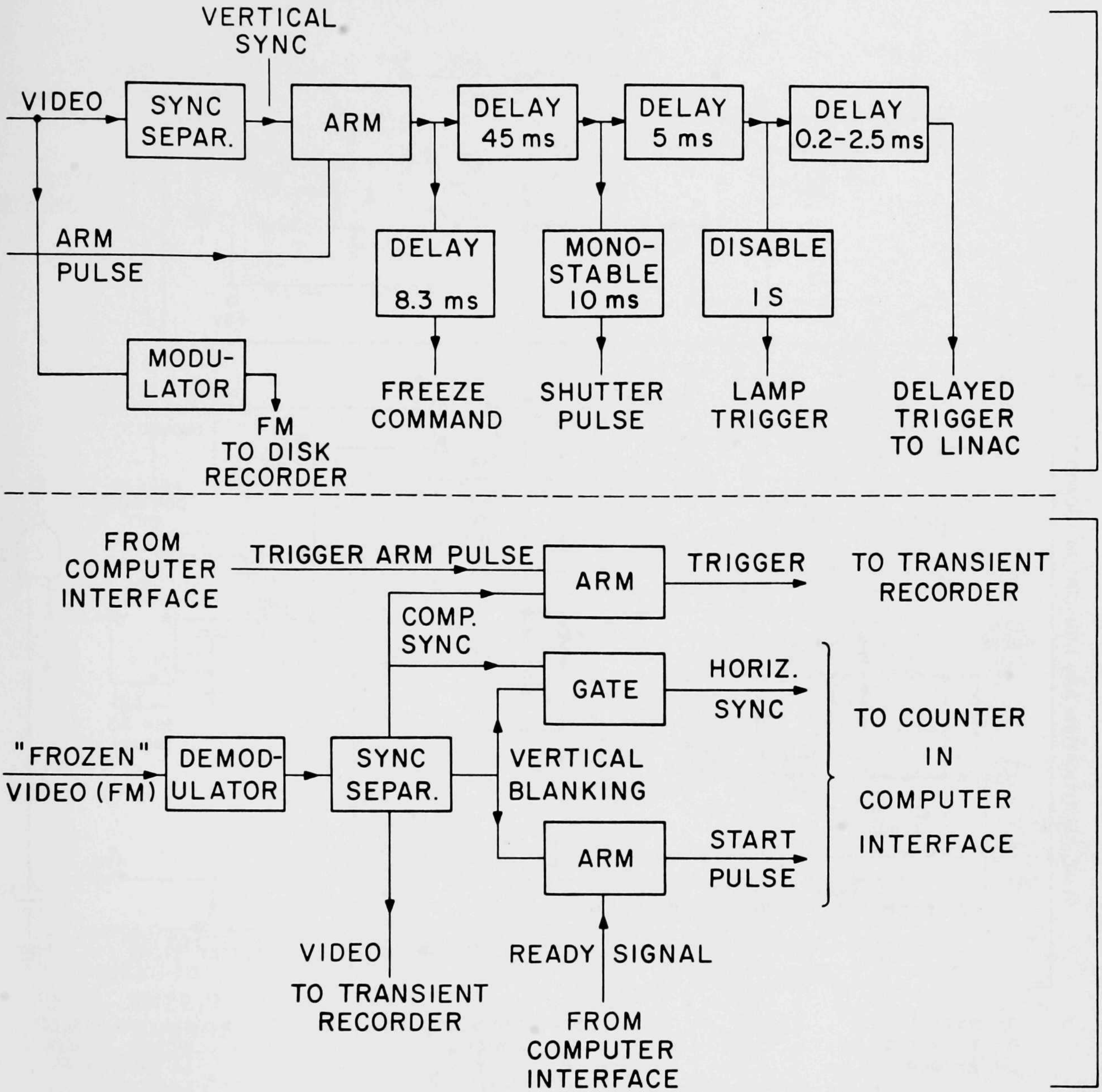


Fig. 4. Detailed Block Diagram of Control Interface Shown in Fig. 2. ANL Neg. No. 122-2654.

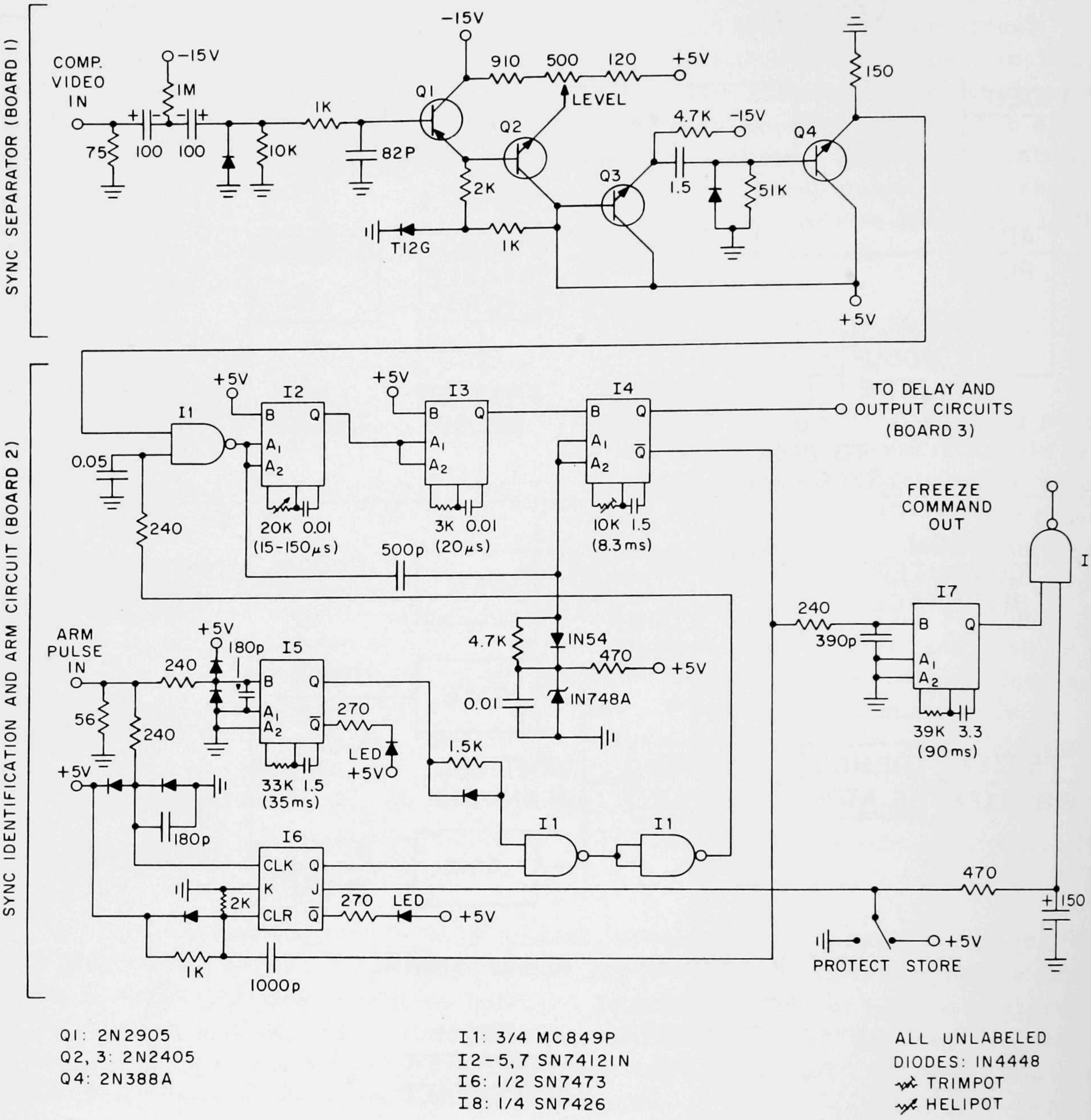


Fig. 5. Sync Separation and Arm Circuits for "Record" Section of Control Interface, with Disk Trigger Output. ANL Neg. No. 122-75-64 Rev. 1.

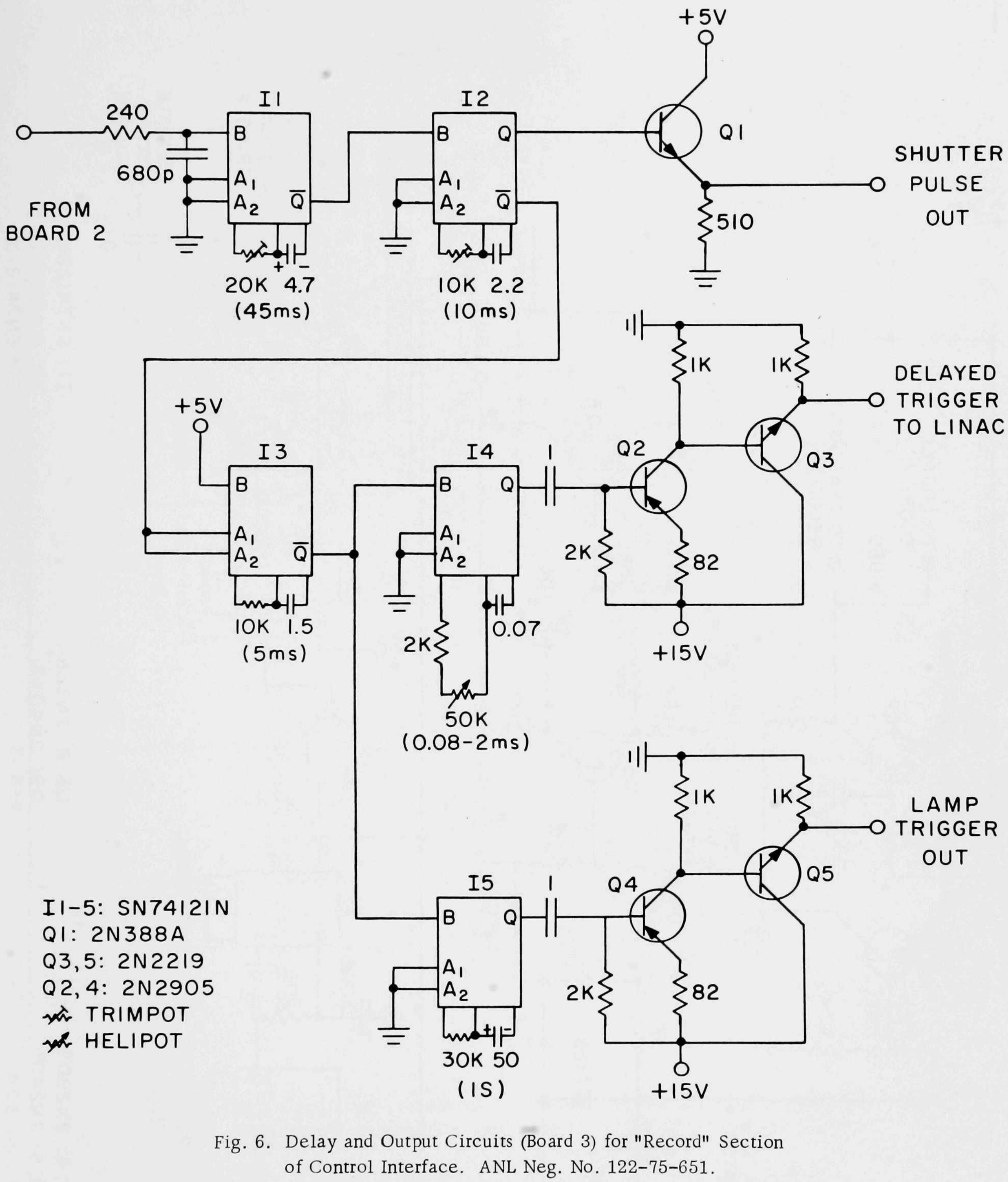
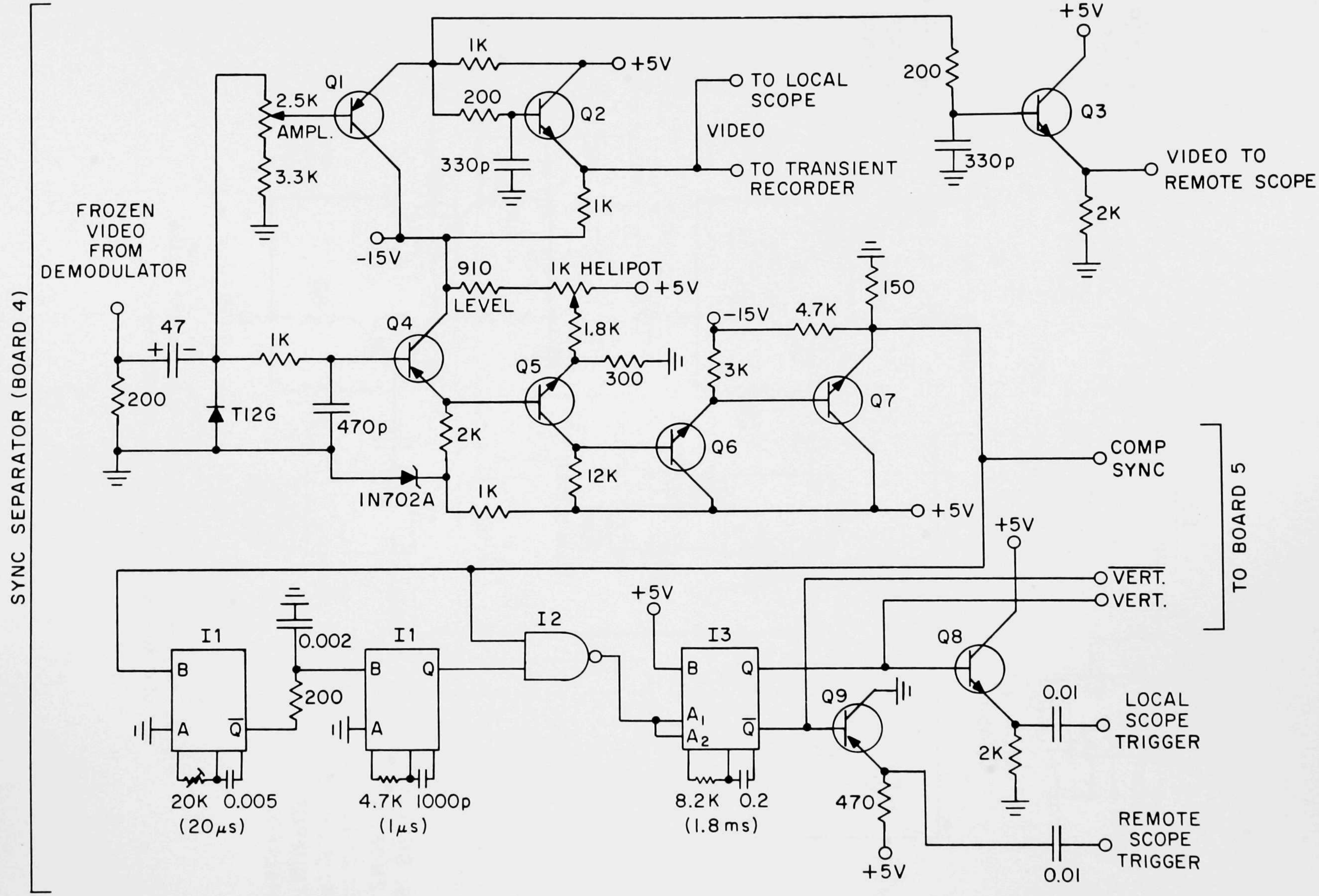


Fig. 6. Delay and Output Circuits (Board 3) for "Record" Section of Control Interface. ANL Neg. No. 122-75-651.



Q1, 4: 2N2905
Q2, 5: 2N2405
Q3: 2N1304

Q6-8: 2N388A
Q9: 2N404A

I1: SN74123N
I2: 1/4 SN7400N
I3: SN74121N

Fig. 7. Sync Separator for "Playback" Section of Control Interface, with Video and Trigger Outputs. ANL Neg. No. 122-75-653 Rev. 1.

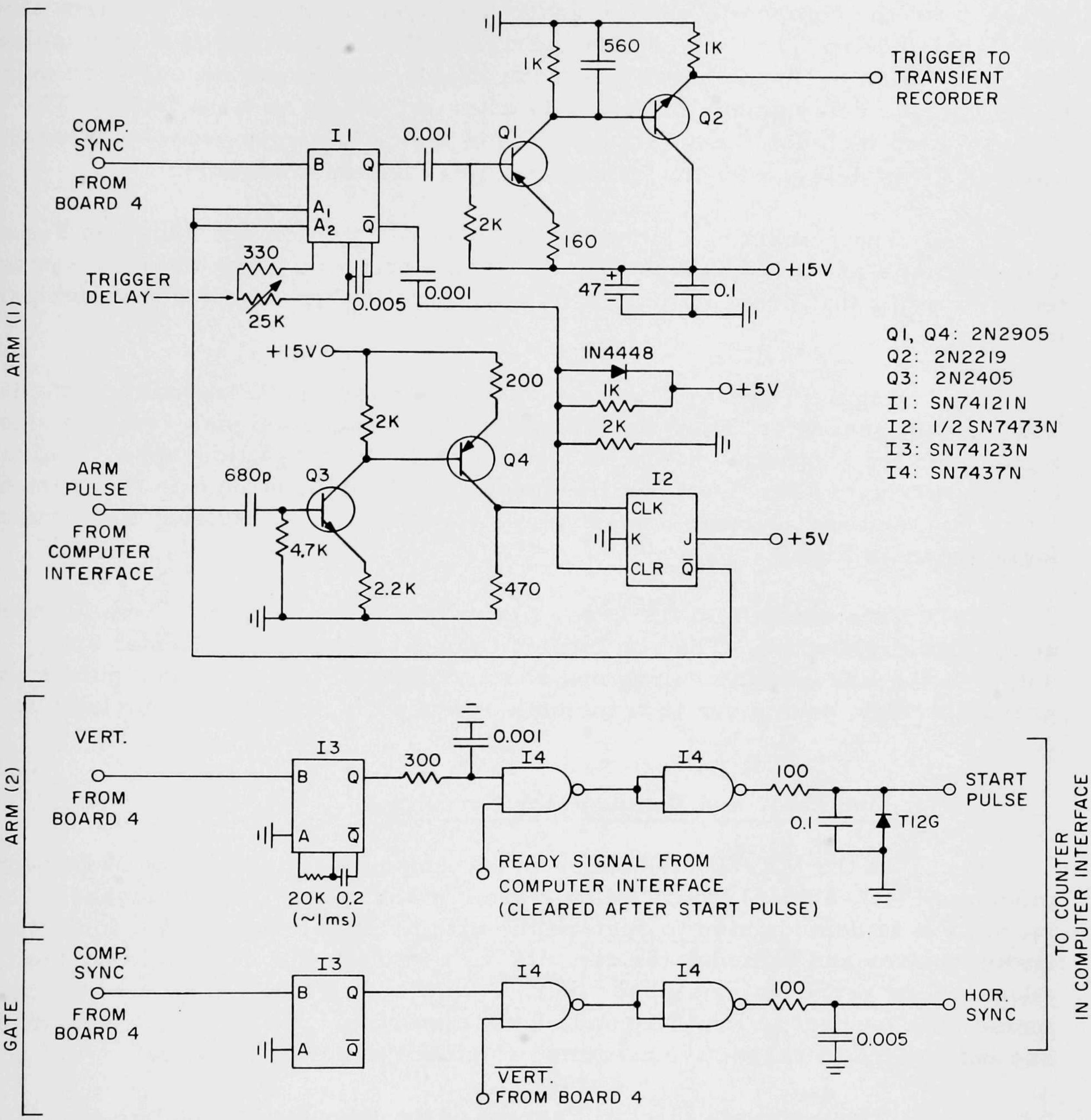


Fig. 8. Logic Controlling Transfer of Stored Streak Picture to Computer (Board 5 of "Playback" section of control interface). ANL Neg. No. 122-75-652.

Diagrams of the first two blocks of the "Record" section are reproduced in Fig. 5. The circuits are practically identical to the corresponding ones described in Ref. 1. The sync separator separates the sync pulses from the composite video signal. The sync identification and arm circuit (labeled "Arm" in Fig. 4, upper part) identifies those vertical sync pulses that precede an even-numbered line scan (field), and transmits one such pulse to the various delay and output circuits after receiving an arm pulse. The diagram also includes the output circuit for the disk trigger, which generates, after an 8-ms delay, a 90-ms, negative-going "freeze command."

The remaining blocks of the "Record" section are shown in Fig. 6. The diagrams are self-explanatory. The lamp trigger output includes a protection circuit that prevents pulses to be transmitted more often than one per second.

Figure 7 contains the sync separator for the "Playback" section. This circuit generates, from the stored composite video signal, isolated video signals for the Biomation 8100 and for two monitoring oscilloscopes. It also furnishes triggers for those oscilloscopes, synchronizing them to the vertical frequency, and vertical and horizontal sync pulses for controlling the remaining logic shown in Fig. 8.

The circuits in the latter figure labeled Arm (1) and Arm (2) need no further explanation. The one labeled Gate disables the horizontal sync output to the line counter during and shortly after the vertical sync pulses to prevent erratic counts due to transients produced by the camera during this time.

4. Modulator and Demodulator Circuits

In the MV-1U disk recorder, the incoming video signal is frequency-modulated (4.4-6 MHz) before being stored on the disk. During playback, the stored FM is demodulated to restore the original video signal. We found that the modulator and demodulator circuits built into the MV-1U, while perfectly adequate for reproducing a good visual TV picture, did not meet our requirements with respect to linearity and signal distortion. We therefore designed and built our own respective circuits, which are described below.

The complete circuit diagram of the frequency modulator is depicted in Fig. 9. The circuit consists of three boards. Board 1 contains a low-pass filter (after Q1), a pre-emphasis circuit (Q2 and Q3), a white peak clipper (after Q3), and a clamp circuit to hold the bottom of the sync pulses at a constant level (after Q4). The circuits on board 1 represent a rearranged and slightly modified version of those used in the MV-1U.[2]

Board 2 contains the modulator circuit proper. It consists of two essentially identical parts (upper and lower halves of the diagram). Each part is composed of a charging circuit (Q1, Q2, Q6, Q7), an integrating

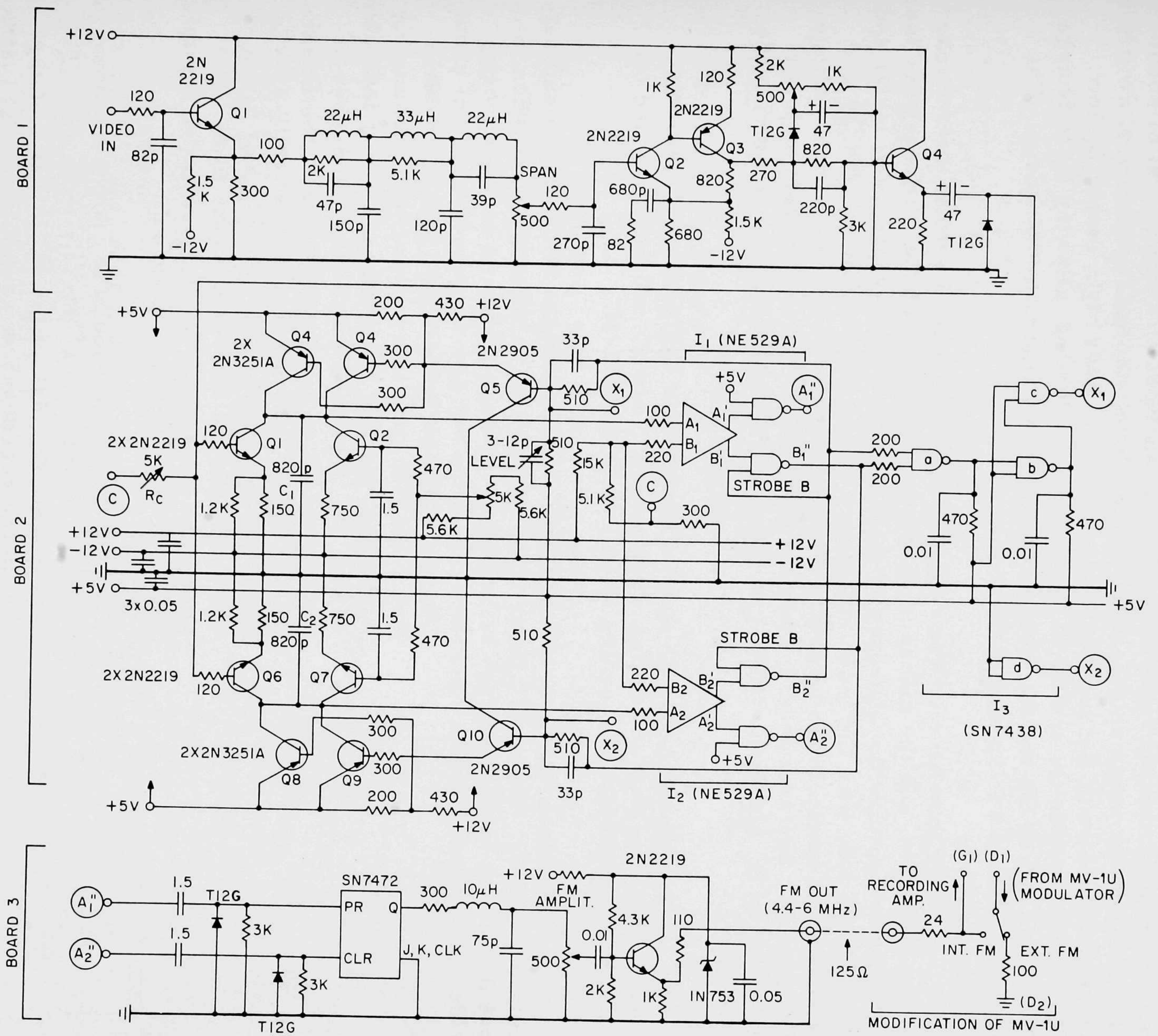


Fig. 9. Circuit Diagram of Frequency Modulator. ANL Neg. No. 122-2649 Rev. 1.

capacitor (C_1, C_2), a discharging circuit (Q3-5, Q8-10), and a fast analog voltage comparator (I_1, I_2). The B output gates of the comparators are wired as a flip-flop. The sequence of events is as follows: Q1 and Q2, acting as current sources, charge C_1 negatively (with respect to the +5 V level). The resulting ramp voltage is applied to input A_1 of I_1. Input B_1 is at a fixed level of about +3.2 V. When the ramp voltage reaches this level, the analog part of I_1 switches, changing the state of the flip-flop; i.e., output B_1'' goes high, output B_2'' low. The former causes Q8 and Q9 to become nonconducting, allowing C_2 to charge. The latter makes Q3 and Q4 conducting so that C_1 begins to discharge. The process continues in an analog manner.

We thus have two sawtooth generators coupled so that their linear ramps alternate, and the discharging time does not affect the frequency[3] as long as it is shorter than the charging time. Since the currents produced by Q1 and Q6 vary linearly with the video signal, and the currents from Q2 and Q7 are constant, the sawtooth frequency is a linear function of the video input voltage. The gates I_3, ac, are used to start the oscillation in the event that both outputs B_1'' and B_2'' are high. Gate I_3, d, acts as a dummy load to symmetrize the circuit.

On board 3, the A'' outputs from I_1 and I_2 alternately preset and clear a flip-flop, thus producing a square-wave signal, which is rounded off and transmitted to the recording amplifier of the MV-1U.

Figure 10 schematically indicates the signals at the inputs and outputs of I_1 and I_2. There is a delay t_d of about 15 ns between the time when input A reaches level B and the beginning of the negative ramp on the opposite side. This should cause a nonlinearity of the voltage-frequency curve of about 3%. As is shown below (Fig. 12A), this deviation from linearity is not observed, presumably due to the "tail" of the discharge curve indicated in Fig. 10. We found a trimmer condenser at the base of Q5 to be useful to "trim" the linearity. It can be shown that a theoretically exact method to compensate for the effect of t_d would be to feed part of the video signal to the B inputs. The trimpot R_c had been intended for this purpose, but was later removed.

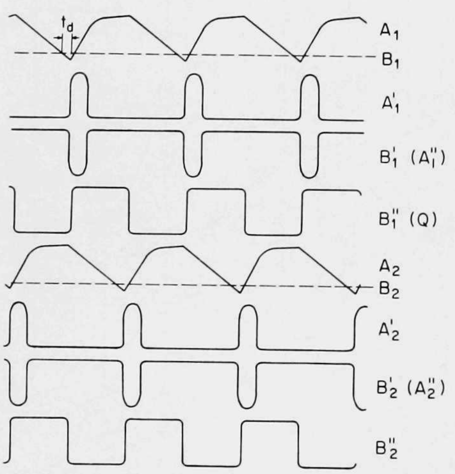


Fig. 10. Signals at Different Points in the Frequency-modular Circuit. ANL Neg. No. 122-2652 Rev. 1.

The demodulator circuit is shown in Fig. 11. It operates in the following way: The signal from the disk of the MV-1U is preamplified in the regenerative amplifier of that instrument and by Q0 and Q1. It is then split into two complementary signals by Transistor Q2. Each signal is transmitted through an emitter follower (Q3, Q4) to the Schmitt trigger input of a monostable (I_1, I_2). The dc levels of the signals are adjusted so that I_1 and I_2 are triggered at the positive and negative zero-crossing points, respectively, of the incoming FM signal. The monostables create negative output pulses of about 50-ns duration, which are converted into current pulses by Q5 and Q6. These current pulses are integrated in C_{int}. The frequency of the current pulses charging C_{int} is twice the FM frequency, and the mean current is proportional to that frequency, i.e., to the original video amplitude.

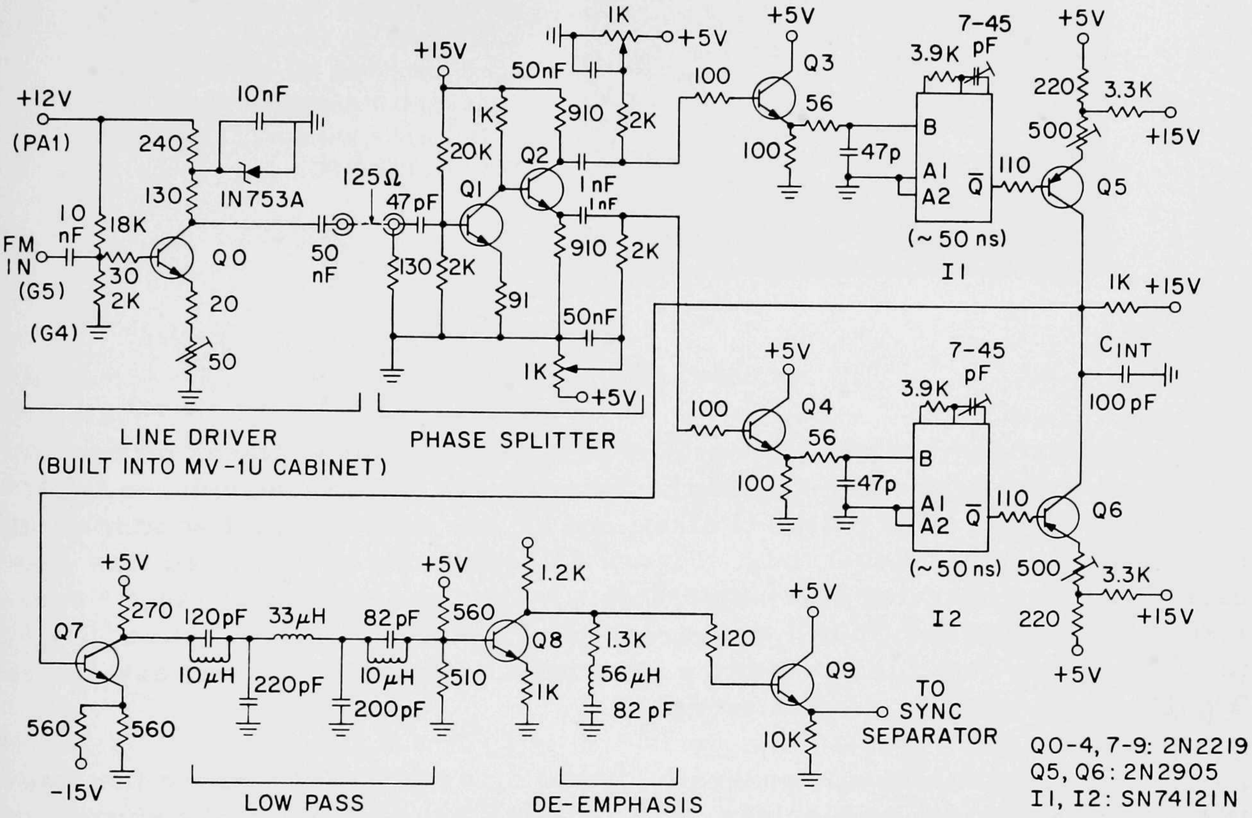


Fig. 11. Circuit Diagram of the Demodulator. ANL Neg. No. 122-2650.

The remaining circuit consists of a low-pass filter (between Q7 and Q8), and a de-emphasis network (between Q8 and Q9). The latter two circuits were essentially copied from the corresponding circuits in the MV-1U.[2]

To test the modulator and demodulator circuits for linearity and signal distortion, the FM output of the former was directly connected to the FM input of the latter and test signals were applied to the video input of the modulator and compared to the signals measured at the video output normally going to the Biomation 8100 (see Fig. 4). Figure 12 reproduces oscillograms of the input and output signals, for two test signals: a sawtooth from an oscilloscope (A) and square pulses (B), with a rise time of 5 ns from a pulse generator. The slight deviation at the bottom of the sawtooth in Fig. 12A is due to the diode clamp circuit in the modulator and a similar circuit in the sync separator. It only affects the lower part of the sync pulses of a video signal. The sawtooth amplitude is 10% larger than the maximum applied video signal.

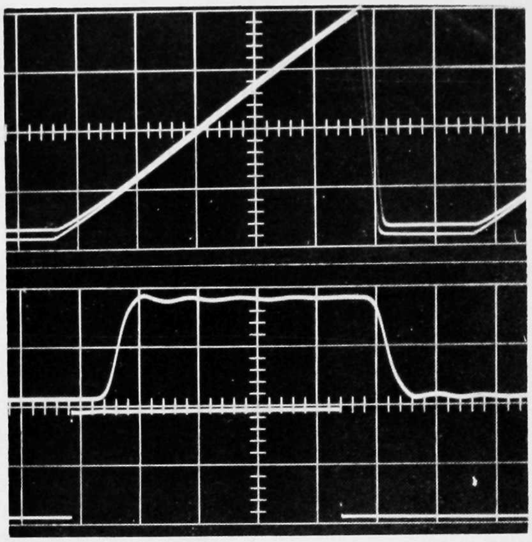


Fig. 12

Linearity and Transient Response of the Modulator–Demodulator System. The lower trace in each picture represents the input signal, the upper trace, the output signal. A: time scale, 10 μs/div; signal amplitude, 1.1 V (10% higher than the maximum range used). B: time scale, 1 μs/div; ordinate 0.2 V/div. ANL Neg. No. 122-2651.

III. COMPUTER PROCESSING

A transient absorption experiment consists of three streak pictures: (1) one with the light source on, but no linac pulse (I_0); (2) one with the light source on and the linac pulsed (Pulse); and (3) one with the light source off and no linac pulse (Dark Level, DL). Figure 13 represents a simplified flow diagram for the processing of the streak data. The data from each "shot," consisting of 2024 values (four-byte words) per TV line, are moved into a temporary raw-data file. A plotting program allows one to display any desired TV line on the graphic display terminal.

A data-compression program, operating on-line, determines the location of the horizontal sync pulses and thus identifies the part of the signal representing the streak picture. It then reduces the useful data from about 800 to about 100 values per TV line by averaging groups of eight or nine values. Also, the blanking level is subtracted from all data to eliminate the effect of signal drift.

The data are subsequently moved into a two-compartment compressed-data file, which has been initially zeroed. In case of a pulse or I_0, the values are added to the respective compartments of the file. For the dark level, they are subtracted from the contents of both compartments. In this way, the sequence of the three shots does not affect the calculation. For each experiment, a separate compressed-data file is opened. This file now contains up to 10000 corrected "pulse" values and the same number of corrected "I_0" values.

The next step is the absorbance calculation. It includes a correction for the nonlinearity (gamma) of the TV camera and the disk recorder circuitry (if any). Also, the portion of the signals before the beginning of the linac pulse is used to correct for slow fluctuations of the light intensity. The program also has the option of calculating emissions (so far uncorrected for the spectral response of the system). In this case, no I_0 is used.

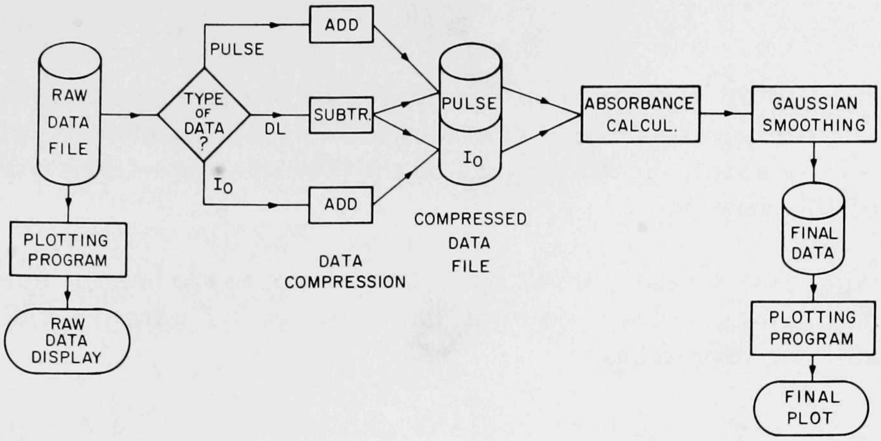


Fig. 13. Flow Diagram for the Computer Processing of the Streak Data (DL = dark level). ANL Neg. No. 122-2648.

The next operation, if so desired, is the smoothing of the data in both dimensions (time and wavelength) by Gaussian convolution. Instructions for the degree of smoothing can be entered at the time of the experiment or at a later time. Absorbance calculation and smoothing are carried out overnight as a "Long Term" (lowest priority) job. The final data (up to 10000 values) are stored in a new file.

All files mentioned so far are disk files. However, the compressed-data files are usually also transferred to tape for archiving purposes.

The final stage of the processing is the plotting on the graphic display terminal. Three plotting programs have been written which produce the following plots: (1) the time function at a desired wavelength, (2) the absorbance spectrum at any given time during the streak, and (3) a three-dimensional plot of absorbance versus time and wavelength. The three-dimensional model can be viewed from any desired angle. Examples of all plots are given in Sec. V.

IV. RANGES COVERED, RESOLUTION, AND ACCURACY

A. General Remark

The values for resolution and accuracy given below are valid when no smoothing of the data is carried out. Furthermore, most of the results shown in Sec. V were obtained before the new frequency modulator had been completed. The absolute accuracy of those absorbance values is therefore two to three times less, and the time resolution two times less than the values reported below.

B. Time

The maximum streak length usable with our system is 2.5 ms. It was obtained by modifying the 10-ms range of a millisecond streak unit. Longer streaks are not possible because part of the TV lines would be scanned before completion of the streak.

The shortest streak length used so far for absorbance measurements is 0.5 μs, a limit imposed by the sensitivity of the TV camera and the radiance of the available light source.

The linearity of the time scale is determined by the linearity of the ramp voltage produced by the streak modules and by the geometrical distortion of the TV camera. We use the following simple method to calibrate the time scale of our system: We repeatedly record the strong transient absorption signal produced in a KCNS solution, varying the setting of the TRW delay generator and thus moving the leading edge of the absorption signal through the length of the streak. The deviation from linearity is typically 1-2% of full scale.

C. Wavelength

With the present system, measurements can be carried out between about 240 and 750 nm, these limits being imposed, respectively, by the loss of uv on the mirrors, and the limited red sensitivity of the S20 cathode of the streak tube. However, we are presently rebuilding the optical arrangement, making it more compact and eliminating all mirrors except those in the spectrograph. By this modification, we expect a considerable intensity gain in the far uv and an extension of the usable range to shorter wavelengths.

With the available gratings of 1200, 600, and 300 lines/mm, the wavelength range covered by one measurement is 85, 170 and 340 nm, respectively. The latter range cannot be fully used in most cases; all wavelengths $\lambda \leq 0.5\lambda_{max}$ have to be eliminated by cutoff filters to avoid second-order spectral components. For measurements in the uv or the red, we usually "straighten out" the I_0 spectrum by reducing the brighter end of the transmitted spectrum with the aid of special filters and/or by slight defocusing, to make better use of the limited dynamic range of the TV detection system.

Since the wavelength range determined by the grating covers 100 TV lines, the wavelength resolution is about 1% of that range, except when the vertical slit width is greater than about 0.2 mm. In the latter case, the resolution is about 5% of the range per mm slit width.

The absolute accuracy of the wavelength measurement is equal to the wavelength resolution.

D. Absorbance

The relative accuracy of the measured absorbance is essentially determined by the noise level observed on the absorbance curve (spectrum or time function), which is composed of three types of noise:

1. Shot noise from the streak tube (ΔA_1).
2. All noise components that can be treated as a noise voltage, including random noise from the Vidicon tube, the video amplifier in the TV camera, and from the entire disk storage system, and "bit noise" from the analog-to-digital converter in the Biomation 8100 (ΔA_2).
3. A nonrandom (with respect to time) component caused by local variations of the gamma of the Vidicon tube (ΔA_3).

An analysis leads to the expression

$$\Delta A = \sqrt{\Delta A_1^2 + \Delta A_2^2 + \Delta A_3^2}, \tag{1}$$

ΔA being the rms noise on the measured absorbance curve, and ΔA_{1-3} corresponding to the three components

$$\Delta A_1 = \frac{\text{const}}{I_0^{3/2}} \sqrt{1 + 10^{3A}}, \tag{2}$$

$$\Delta A_2 = \frac{1}{\gamma \, \ell n \, 10} \frac{\Delta V}{V_0} \sqrt{1 + 10^{2\gamma A}}, \tag{3}$$

and

$$\Delta A_3 = \frac{A}{\gamma} \Delta\gamma. \tag{4}$$

In Eqs. 2-4, I_0 is the light intensity at a given wavelength, incident on the photocathode of the streak tube, A is the absorbance calculated from the experimental data, γ is the gamma value of the Vidicon tube as defined by Eq. 5, ΔV is the rms noise voltage, and V_0 is the signal amplitude corresponding to I_0. ΔV and V_0 are measured at the input of the Biomation 8100. The relation between light intensity and measured voltage is

$$V = \text{constant } I^{\gamma}. \tag{5}$$

We found that, under optimum conditions (measurements in the near uv or the lower part of the visible spectrum), the component ΔA_1 is negligible. For our present Vidicon ($\gamma = 0.7$), we calculated from recorded dark-level signals the minimum obtainable ΔA as

$$\Delta A_{min} \approx \Delta A_2 = 0.0026\sqrt{1 + 10^{1.4A}} \tag{3a}$$

or, for $A \approx 0$, $A_{min} = 0.0037$.

This value is in approximate agreement with the lowest noise level observed on measured absorbance curves. Under more adverse conditions (measurements in the far uv or the red, or on strong absorbing solutions), the first component becomes important, and the noise level typically increases by a factor of 2-4. The third component only affects measurements of relatively high absorbance values.

The fact that the first two components, ΔA_1 and ΔA_2, increase rapidly with the absorbance, imposes a practical limit to the measurable absorbance: Values over 1 cannot be determined with reasonable accuracy.

The absolute accuracy of the absorbance depends on how well the response curve of the system consisting of the TV camera and the video and disk circuitry can be approximated by a correction function incorporated into the computer program (such as Eq. 5). Again, local variations of the response curve of the TV camera play an important role in this respect.

From experiments with neutral-density filters, we conclude that the absolute error of a transient absorbance measurement, not counting the noise-related error, is normally less than 3%.

V. RESULTS

Figure 14 shows three-dimensional plots of a transient spectrum produced by a pulse of electrons in an aqueous solution of p-xylene. The two representations (A and B) result from rotation of the three-dimensional representation relative to the projection axes. To reveal the important features of a transient spectrum more clearly, any set of angles may be selected as an option in the FORTRAN plotting program. In our example, one can see two major peaks (due to H and OH attack) decaying at different rates. Figures 15 and 16 illustrate the spectrum of this same transient at two different times after the pulse. Here again, one can see that one of the major peaks decays more rapidly than the other. In Figs. 17 and 18, we show still another option in displaying the data. Here we illustrate the decay of the transient at two different wavelengths. From these decay curves, it is possible to obtain rate constants by appropriate FORTRAN programs.

Figure 19 shows a three-dimensional display of the emission spectrum produced by irradiating a solution of naphthalene in benzene with a 10-ns pulse of electrons.

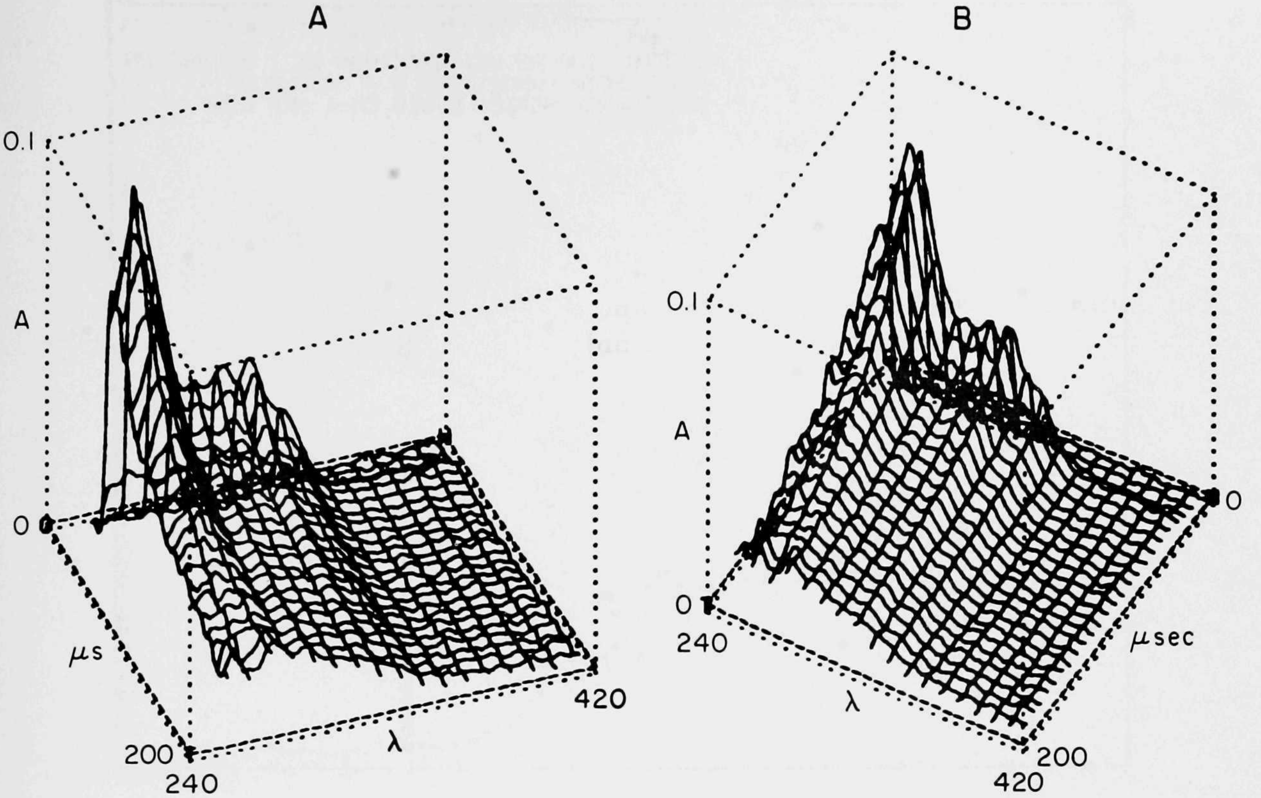


Fig. 14. Three-dimensional Plots of Transient Absorption Spectrum Observed in Pulse-irradiated p-xylene. ANL Neg. No. 122-2653.

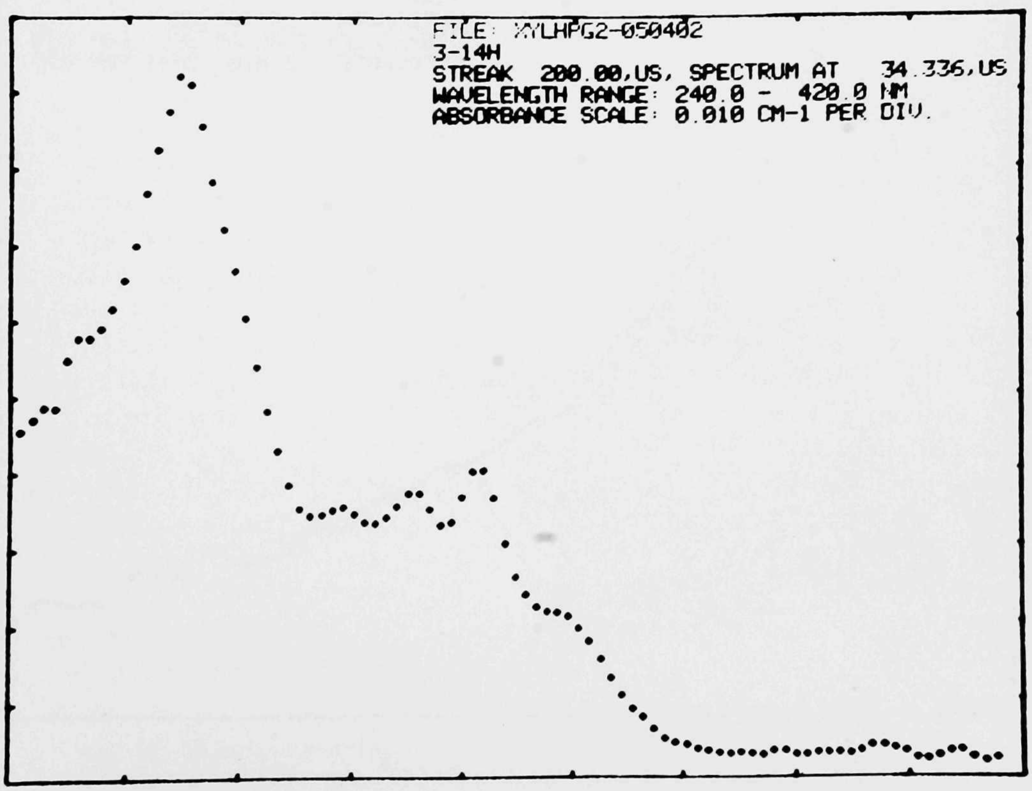


Fig. 15. Spectrum of p-xylene Transient Immediately after Electron Pulse. ANL Neg. No. 122-1239.

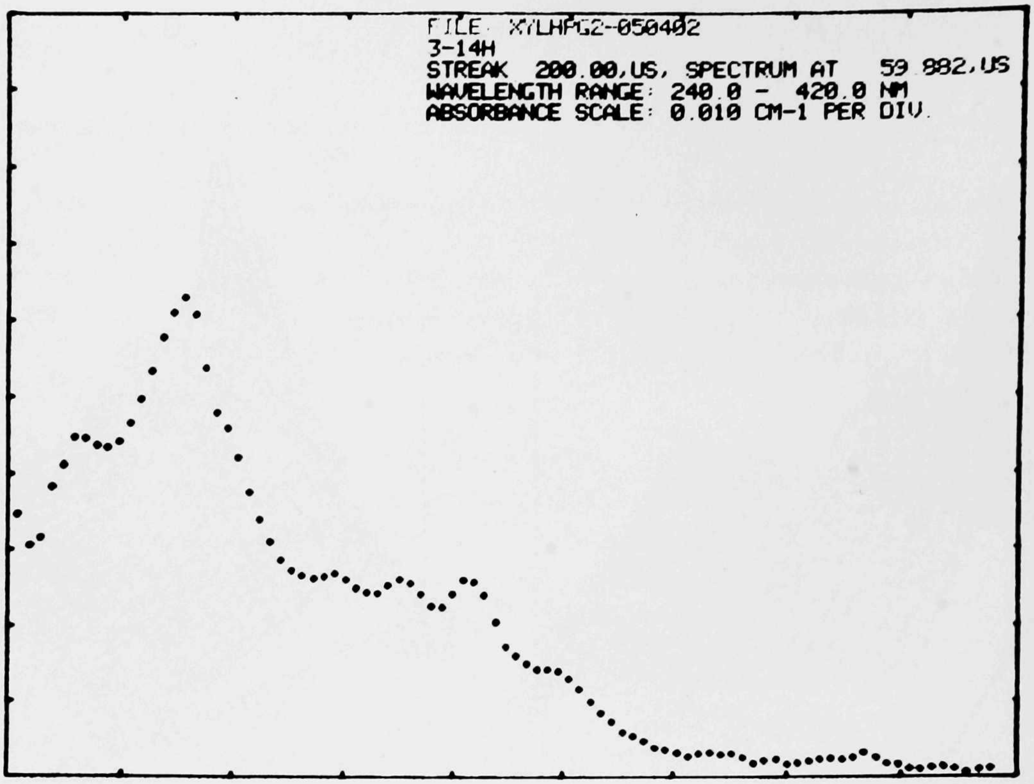


Fig. 16. Spectrum of p-xylene Transient 27 μs after Electron Pulse. ANL Neg. No. 122-1236.

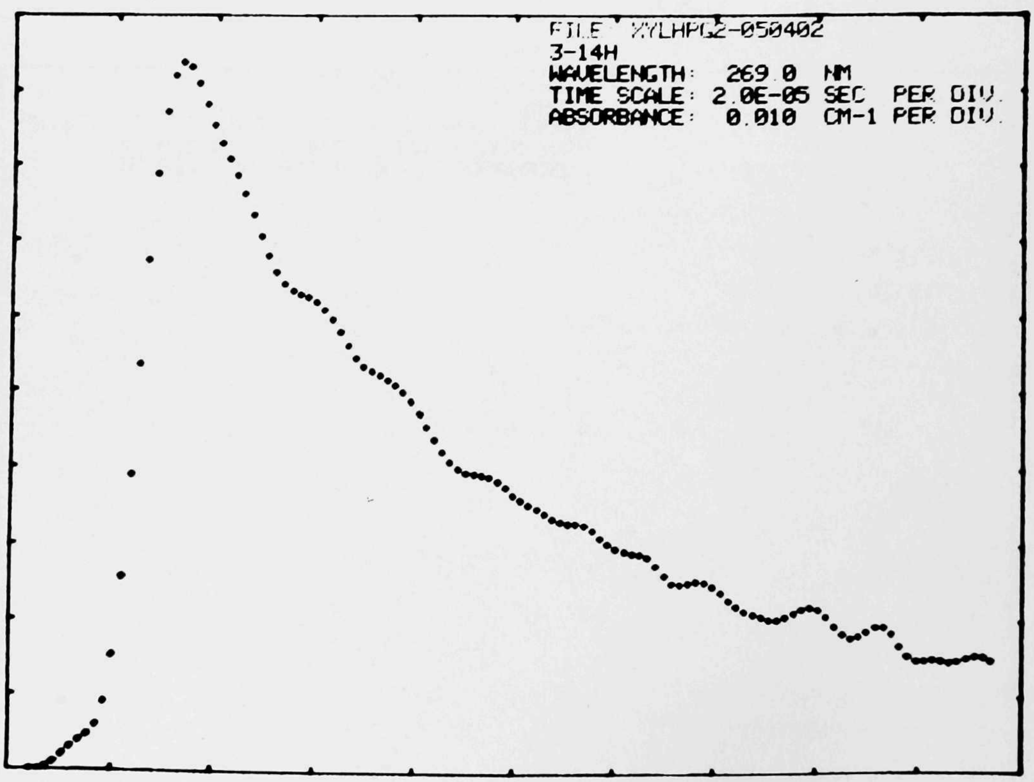


Fig. 17. Time function of p-xylene Transient at 269 nm. ANL Neg. No. 122-1238.

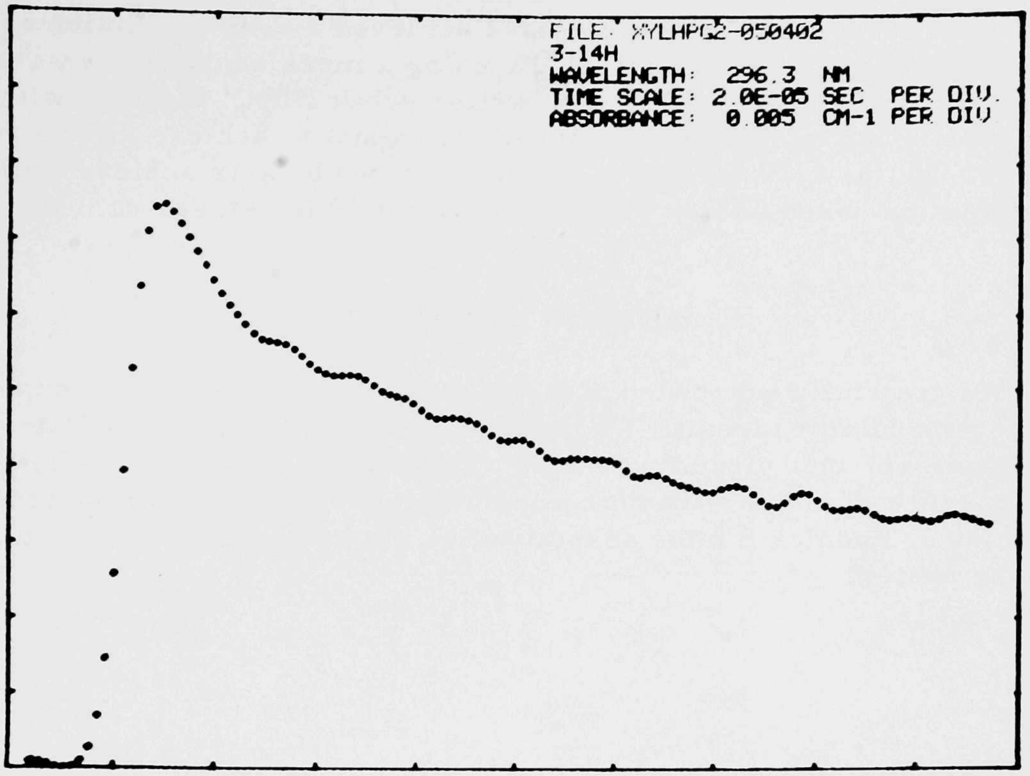


Fig. 18. Time function of p-xylene Transient at 296 nm. ANL Neg. No. 122-1235.

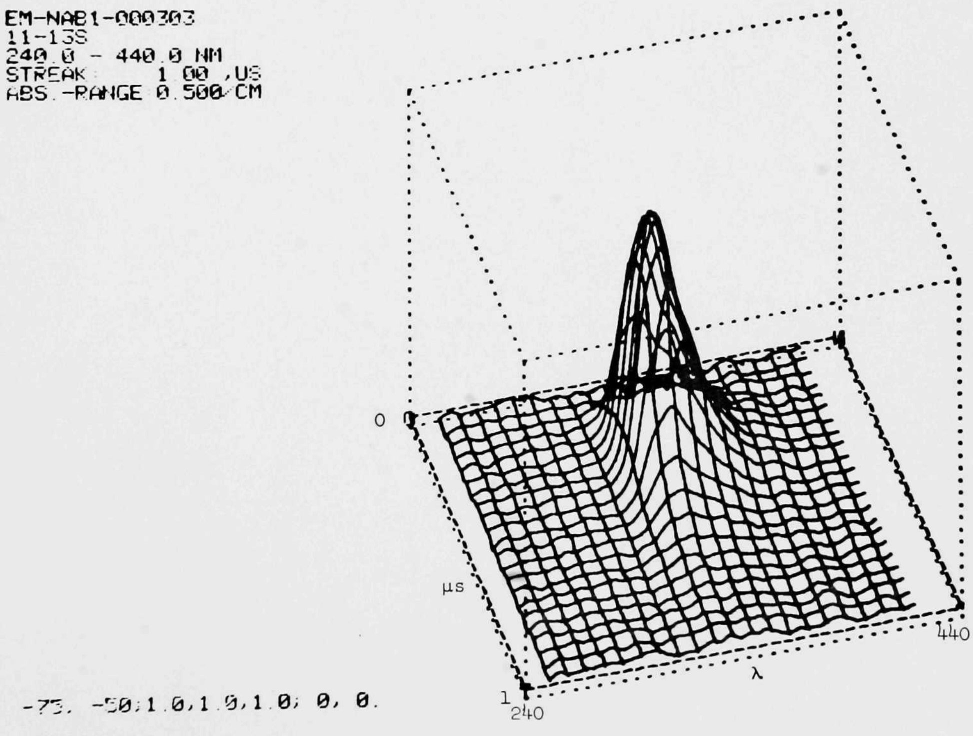


Fig. 19. Transient Emission Produced in a Solution of Naphthalene in Benzene Irradiated with a 10-ns Electron Pulse. (Time range, 1 μs; wavelength, 240–420 nm; emission in arbitrary units.) ANL Neg. No. 122-2619.

With the present system we have achieved a time resolution of 10 ns in absorption and 1 ns in emission. By using a more sensitive camera of the ISIT type which we described in our earlier publication,[1] together with a faster streak module for our present camera,[4] we expect to achieve a time resolution of 200-500 ps in absorption. In emission, we hope to achieve still shorter time resolution based on our experience with a faster streak camera.[5]

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